COO-1156-117

RADIOECOLOGY OF NATURAL SYSTEMS

FINAL REPORT

for the period

May 1, 1962 - October 31, 1979

1972-?

F. W. Whicker, Principal Investigator

Department of Radiology and Radiation Biology

Colorado State University

Fort Collins, Colorado 80523

October 31, 1979

prepared for

The U. S. Department of Energy

under Contract No. EY-76-S-02-1156

VI. PLUTONIUM BEHAVIOR IN A GRASSLAND ECOSYSTEM

During the summer of 1972, pilot studies on the distribution, transport, and ecological effects of plutonium in the terrestiral environs of the Rocky Flats nuclear weapons facility near Denver, Colorado were initiated. Because of uncertainties in funding however, the work did not gain much momentum until 1973. By mid-1974, this program was receiving the majority of the effort under the contract and this trend has continued through October 1979.

The Rocky Flats facility, presently operated for the Department of Energy by Rockwell International, has handled large amounts of plutonium for defense purposes since approximately 1950. During the period 1959-1964, drums containing plutonium-contaminated cutting oil were deteriorating and leaking onto the ground of the former barrel storage area. During this period, and until 1969 when all barrels had been removed and the contaminated area had been covered with asphalt, the barrel storage area was a source of contamination of the surrounding area. Physical disturbance of the area and resuspension of contaminated soil by winds are primarily responsible for the initial distributions of plutonium to the environs. Other accidental events and routine releases also caused some contamination of the environs, but these sources were minor in comparison to the barrel storage area. Most of the Pu contamination was deposited to the east and southeast of the source area because of the prevailing directions of the strong winds. Public concern, a lack of knowledge on the environmental behavior of Pu, and its high radiotoxicity all provided justification and impetus for this investigation.

A. Ecosystem Distribution

To determine the distribution of Pu in the terrestrial environment, several sampling areas were established. Three areas were near and downwind

2

of the source: a 0.25 ha plot 50 m southeast, a 0.75 ha plot 200 m southeast, and a 500 m transect extending from 270 to 770 m southeast. Two other study plots were established; one 1400 m south of the source area, and one in the northwest corner and upwind of the plant. During 1972-1976, samples of soil, litter, vegetation, arthropods, snakes, birds, small mammals, and mule deer were taken from the study plots for Pu analysis and biomass estimates. Samples were analyzed by liquid scintillation counting in our laboratory and by alpha spectrometry by commerical laboratories.

Based on data from the principal study plot 200 m downwind of the source area, the top 21 cm of soil contained over 99% of the ecosystem Pu inventory. About half of this was in the upper 3 cm. Fractions of the total inventory in non-soil compartments were orders of magnitude lower than the soil inventory. The inventory exhibited the following ranking: soil >> litter > vegetation >> arthropods > small mammals > other animals. A major reason for soil so-dominating the Pu inventory is the comparatively insignificant mass/unit area of the other compartments. Although soil contained the highest concentrations of Pu of the materials studied, concentration ratios were reasonably high, especially for litter and vegetation. However, much of this result was due to surficial dust that was not removed from the litter and vegetation prior to Pu assay. Concentration ratios relative to 0-3 cm soil ranked as follows: litter > vegetation > arthropods > small mammals > other animals. While Pu levels in litter, vegetation and arthropods were always readily detectable, levels in small mammals were occasionally non-detectable and levels in other animal tissues were seldom detectable.

3

Plutonium concentrations in soil varied inversely with distance from the source area, depth of sample, and particle size of dry-sieved samples. Coefficients of variation of Pu in soil samples randomly collected within sampling grids ranged to more than 300%, and frequency distributions were highly skewed with most values lying below the mean and a few being much larger than the mean. The measured plutonium distribution patterns and known characteristics of the source imply the following dispersal mechanisms: (1) the attachment of PuO_2 to soil particles; (2) primary dissemination of the contaminated soil particles by disturbance and wind; and (3) weathering of the contaminated soil particles. The high degree of spatial variability suggests that the most common functional form of the contaminated soil during dissemination was an agglomerated particle containing a large number of individual grains of soil and PuO_2.

In an effort to more fully understand the problem of sampling heterogeniety, soil samples from the principal study plot were examined microscopically so that plutonium particle sizes and micro-distribution could be investigated. We wanted to explore the idea that occasional high-activity samples were possibly caused by "hot particles" of plutonium. An autoradiographic technique, utilizing nuclear emulsion plates, was used to obtain abundance of, and equivalent size distributions for, plutonium particles in Rocky Flats surface soil. A total of 1700 particles were sized, for which mean $^{239}PuO_2$ equivalent diameters of 0.29 μm, 0.25 μm, and 0.20 μm were found for 7-, 14-, and 37-day exposures of soil to emulsion plates. A method to scan for particles greater than 1.50 μm equivalent diameter, utilizing Kodak AA Industrial X-Ray film, was also used. The largest particle sized with this procedure was 6.86 μm $^{239}PuO_2$ equivalent diameter. None of the particles sized was of sufficient magnitude alone to account for elevated plutonium activity observed in Rocky Flats surface soil samples. Variability in particle concentrations was observed, however, which suggested that heterogeneity in the spatial distribution of plutonium particles in the soil may partially explain observed variability in soil plutonium concentrations. While no "hot particles" (> 1000 dpm) were found, their presence cannot be ruled out because of the limited quantity of soil that can be feasibly examined by the autoradiographic technique.

Based upon data from samples taken across gradients of plutonium contamination, levels of plutonium in litter, vegetation and arthropods were significantly correlated to concentrations in soil. These correlations provided additional evidence that most of the plutonium associated with these samples was actually attached to surficial dust. This study corroborates the idea expressed by other investigators that plutonium moves in the environment principally by physical rather than physiological mechanisms.

Data on ^{239}Pu concentrations in vegetation, small mammals and arthropods were too variable to show statistically significant differences between species or taxonomic groups. It also could not be shown that there were significant differences between collection dates nor types of small mammal tissues. Statistical analyses were exhaustive in that data were log-transformed to reduce heterogenous variance and skewness of distributions, and normal as well as non-parametric procedures were used. It is concluded that the large variability in the data are real, and not the result of poor sampling and/or analytical technique. This kind of variability is probably representative of that likely to be encountered from accidents or single events. Smaller sampling variability for world-wide fallout and routine low-level discharges are more likely because of smaller particle sizes and more opportunity for stochastic mixing processes.

5

Samples analyzed by alpha spectrometry provided data on the isotopes ^{239}Pu and ^{238}Pu, and thus the opportunity to examine their comparative behavior was afforded. The activity isotopic ratio of ^{239}Pu to ^{238}Pu (IR) for surface soil was about 65. The IR was found to decrease with depth, and IR values for many animal tissues were also substantially lower. The initial interpretation of this observation was that ^{238}Pu must be more mobile than ^{239}Pu. This concept has some experimental basis if the two isotopes occur in separate particles. However, there is no basis for thinking this to be the case in the contamination at Rocky Flats. Individual particles of PuO_2 likely contain both isotopes, and furthermore, there is considerable evidence for monomeric Pu in environmental samples. Further examination of the data indicates that the IR is biased on the basis of total Pu activity present. Above 10 dpm, the IR appears independent of sample activity, but below that value, the IR decreases. This suggests some low-level interference in the alpha peak of ^{238}Pu, and our present hypothesis is that the IR effect is basically an analytical problem.

B. Transport Processes

In an effort to understand the dynamic behavior of plutonium in the environs of Rocky Flats, several studies dealing with transport processes were undertaken. These investigations included measurements of aerial deposition rates, soil erosion, plutonium export initiated by the activities of pocket gophers and mule deer, and mechanisms of plutonium accumulation by vegetation. The construction of a Pu transport model is presently possible, based on the results of these studies, and those of other laboratories at Rocky Flats. However, this has not been accomplished because other efforts have taken priority.

6

In 1976 and 1977, aerial deposition rates were measured in the principal study plot (macroplot 1) with 21 wet pot collectors. The data were in reasonable agreement with the Rockwell air monitoring data, but there was some indication that conventional deposition from the air column was not the only mechanism involved. Dust generated by vehicular traffic and rain splash-up of surface particles were probably more important mechanisms of deposition in the pot collectors.

Since over 99% of the plutonium in the ecosystem is in soil, erosional processes, both wind and water driven, could disperse significant quantities of the element. These processes can be enhanced considerably by natural phenomena such as animal activity and needle ice formation, and by human disturbance. Attempts were made to measure soil erosion by tracing the location of ^{59}Fe-tagged soil particles and by recording beta counts from sealed ^{90}Sr sources buried at fixed positions beneath the soil surface. These very sensitive techniques gave no indication of measureable soil erosion over the duration of the study in undisturbed, well-vegetated areas. However, in steep or disturbed areas, evidence of erosion was visually observed but not quantified.

The potential of certain animals for dispersing plutonium was investigated in detail for pocket gophers and mule deer, and superficially for arthropods. For example, pocket gophers at Rocky Flats dig tunnels and burrows and form mounds which expose contaminated soil to erosional processes. Quantitative estimates of the amount of soil thus exposed per unit area per year, and the amounts of Pu in mound soil were made. Since most of the mound soil originates from a depth of 10-15 cm, it contains less Pu than the surface soil. The fate of plutonium in exposed mounds was not quantified, but observations indicate that most of the mound soil

1

and the Pu contained in it remains within a few meters of the original mound. Mule deer ingest plutonium associated with vegetation and soil and transport the material to the surrounding area where most of it is deposited on the ground in fecal pellets. It was estimated that deer transport less than 10^{-7} of the Pu inventory annually, and that most of this is redeposited on DOE-controlled property. Observations indicate that the most heavily contaminated area is utilized only occasionally by deer. Arthropods, because they harbor a larger inventory of Pu than other animals, and because many of them are very mobile and have rapid population turnover rates, probably export considerably more Pu to the surroundings than other animals. However, data indicate that total biological export is very small and probably on the order of 0.001% per decade. Of this, less than 10% is likely to leave DOE-controlled property. In time, plutonium is slowly migrating to deeper soil layers. Thus, it is expected that export, both physical and biological, will diminish through time, assuming that the buffer zone around the plant is protected from disturbance and a good cover of natural vegetation is maintained.

Based on greenhouse studies, transplanted sod blocks in the field at Rocky Flats, and comparison of Pu content of ultrasonicated to unwashed vegetation in field plots, it is concluded that aerial transport and surficial contamination dominates substantially over root uptake from soil. Of the plutonium associated with vegetation, over 90% appears to be surficial, which accounts for the comparatively high concentration ratios obtained in the inventory and distribution phases of the program. However, the point is made that evaluation of plutonium ingestion by herbivores must account for such surficial deposition. Further, such material is of importance from the standpoint of export and possible biological effects.

As plutonium migrates into the soil and as litter and uncontaminated dust accumulate on the soil surface, the aerial transport mechanism for the present Pu inventory should diminish.

C. Ecological Effects

The contamination of a portion of the terrestrial environs of Rocky Flats with substantial quantities of plutonium provided the opprotunity to search for possible ecological effects resulting from the presence of the element. Gross population effects from the levels of plutonium present in the environment were not expected on the basis of simple dose calculations and data from laboratory studies. However, prior to our studies, a lack of population effects had not been demonstrated and furthermore, extrapolation from the laboratory to the natural environment is frequently not a valid practice, owing to complexities and interactions which occur in nature but not in the laboratory.

We conducted studies which permitted comparisons of various biological measurements and pathological data between ecologically similar study areas at Rocky Flats of widely varying plutonium levels. Soil in the principal study areas ranged from 100 to over 20,000 dpm ^{239}Pu/g in the upper 3 cm (2-400 $\mu Ci/m^2$). In addition, comparative data were obtained from control areas, containing only world-wide fallout plutonium of the order of 0.1 dpm/g (0.002 $\mu Ci/m^2$). Biological measurements such as vegetation community structure and biomass; litter mass; arthropod community structure and biomass; and small mammal species occurrence, population density, biomass, reproduction, and physical size of whole carcass and organs were made. In addition, pathological examinations of small mammals, including x-ray for skeletal sarcomas, microscopy for lung tumors, and necropsy for general pathology and parasite occurrence were carried out.

While minor differences in certain biological attributes between study areas were observed, none could be related to plutonium levels. Pathological conditions and parasites were found in some rodents, but occurrence frequencies between control and contaminated areas were similar. No evidence of cancers or other radiogenic diseases was found. These observations and measurements, combined with intensive field observations over a period of five years, led to the conclusion that plutonium contamination at Rocky Flats has not produced demonstrable ecological changes. Furthermore, the levels of plutonium observed in tissues of plants and animals in contaminated areas were insufficient to produce the doses that would be required to produce obvious biological changes. Subcellular biological changes, such as chromosome aberrations, cannot be ruled out at Rocky Flats. However, even if chromosome aberration frequencies were increased in the more highly contaminated areas, population-level changes would likely not persist because of the surrounding reservoir of normal genetic information, and because of natural selection.

Based on all the plutonium work we conducted in the terrestrial environs of Rocky Flats, there is strong evidence that the element is not likely to pose an ecological hazard unless extremely high levels (>>1 mCi/m^2) occur. The major reason for this is the extremely low biological mobility of the common chemical forms of the element, amply demonstrated in this and other research. Although uncertainty exists as to possible long-term changes in biological availability of plutonium, we expect gradual soil penetration and dispersion to diminish the present hazard potential with time. Furthermore, if natural thorium is a reasonable geochemical analogue of plutonium as many scientists believe, then the extremely low biological mobility of primordial thorium should be generally indicative of the long term mobility of plutonium.

10 | 10